Velocity distributions in microskimmer supersonic expansion helium beams: high precision measurements and modelling

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Supersonic molecular beams are used in many applications ranging from spectroscopy and matter wave optics to surface science. The experimental setup includes a conically shaped, circular aperture, popularly referred to as the skimmer which is used to collimate the beam. The skimmer diameter typically ranges between a few hundred μm and a couple of mm. Recent years have seen an increased use of so called microskimmers. It has been reported that microskimmers with diameters below 10 μm produce beams with significantly broader velocity distributions (smaller speed ratios) than standard skimmers. Various explanations for this phenomenon have been proposed, but up till now only a limited amount of data has been available. Here we present a systematic study of the velocity distribution in microskimmer supersonic expansion helium beams. We use a source design which allows sub-micrometer precision positioning of the skimmer relative to the nozzle. The velocity distributions have been determined with high precision using a modified method we have recently developed. We compare the measurements of a 4 μm diameter skimmer with measurements of a 390 μm diameter skimmer for room temperature and cooled beams in the pressure range of 11 bar to 181 bar. Our measurements show that for properly aligned skimmers, with a sufficiently large opening angle, there is no difference in the velocity distribution. The only difference is that the most probable velocity for a given pressure and temperature is slightly lower for the microskimmed beam. We ascribe this to the higher knudsen number for the microskimmers. We fit our measurements with a model for the supersonic expansion and obtain good agreement between the experiments and simulations.

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I. INTRODUCTION

Supersonic molecular beams are used in a range of scientific disciplines. Helium beams in particular are an established tool in surface science used in diffraction experiments, dynamics studies (diffusion and surface vibrations) and for monitoring thin film growth and thermal evaporation[1–10]. Work is ongoing to extend the use of helium beams to direct imaging in neutral helium microscopes[11–15]. Molecular beams can also be employed as a carrier gas for deposition of other molecules[16].

A supersonic molecular beam is created by a supersonic (free jet) expansion: atoms or molecules from a high pressure reservoir (typically up to 200 bar or more) expand into vacuum through a nozzle with a diameter larger than the mean free path of the gas particles in the reservoir. The expansion is adiabatic. As the atoms or molecules expand into vacuum they collide until free molecular flow is reached. The advantage of the supersonic expansion compared to an effusive beam is the high beam density and narrow velocity distribution that can be achieved[17]. The central part of the beam is selected by a conically shaped, circular aperture, popularly referred to as the skimmer.

For most experiments the skimmer has a diameter between 200 µm and a few mm. The first experiments using a microskimmer were presented by Braun et al.[18]. This paper introduces the method of glass pulling for the creation of microskimmers which is used to this day. Measurements were obtained using a source pressure of 120 bar and a 10 µm diameter nozzle. In the paper it is reported that speed ratios for 3 µm and 5 µm skimmers are considerably lower than those for a standard 1.6 mm diameter skimmer: 65 and 24 respectively compared to 78 for the standard skimmer. The speed ratio is a standard way to express the quality of a molecular beam and is defined as $2ln2u/\Delta u$ where u is the most probable (mean) velocity and Δu is the full width at half maximum of the velocity distribution[19,20].

Braun et al. propose geometrical imperfections and/or imperfections of the lip edge of the skimmer as well as difficulties in aligning the skimmer and nozzle as possible explanations for the lower speed ratios. In their paper they suggest that microskimmers can be used for atom optics experiments and indeed up till now this has been the main application. The first experiment using a microskimmer to focus a neutral helium beam was carried out by Doak et al.[21]. Focussing measurements were carried out using skimmers between 1 µm and 14 µm in diameter with a source pressure up to 150 bar and a 5 µm diameter nozzle. The expected focussed spot diameter was not achieved. The relative deviation between expected and measured focus increases from 1.1 for a 14 μm skimmer to 55 for the 1 μm skimmer. It is suggested in the paper that this is due to the supersonic expansion continuing after the beam has passed through the skimmer aperture. It is stated that measurements were carried out for velocity distributions between around 1 % and up to around 10 % (corresponding to speed ratios between around 140 and 14). These speed ratios are not compared explicitly to standard skimmer measurements. The first neutral helium microscopy images were obtained a few years later. The resolution was around 2 μm, using a 1.2 μm diameter skimmer[11]. Experiments were also carried out with a 2.4 μm diameter skimmer. The paper states that speed ratios between 16 ± 1 and 140 ± 3 were obtained with source pressures between 11 bar and 191 bar using a 10 μm diameter nozzle. The paper also states that chromatic abberations caused by the velocity distribution of the beam is the resolution limiting factor and that no signs of further expansion after the beam has passed through the skimmer could be observed. The first sub-micrometer focussing was obtained by Eder et al.[12]. A mikroskimmer 1.1 μm in diameter was used. The measurements were performed at a source pressure of 81 bar and 110 bar using a 10 μm nozzle. However the velocity distributions were not measured explicitly, instead theoretical values were used to calculate the expected focus size. The agreement was good, but the measurements had large error bars.

The importance of the speed ratio for the microscope resolution is discussed in[22]. This, together with the discussion above illustrates how important it is to determine the true, best obtainable velocity distribution from microskimmers. In this paper we present such a detailed study. Of particular importance is the use of our molecular beam source which allows the skimmer to be positioned with sub-micrometer precision relative to the nozzle[23]. Microskimmer measurements are compared with measurements using a standard skimmer and we are using a new method which we have recently developed which enables us to measure an accurate velocity distribution from our TOF measurements even for high speed ratios[24]. Further we use our theoretical model for the supersonic expansion described in[25,26] to model the experimental data. The model is described in section 3.

II. EXPERIMENTAL SETUP

The experiments presented here were carried out in the molecular beam apparatus at the University of Bergen, popularly referred to as MAGIE[27]. A drawing of the experimental setup can be

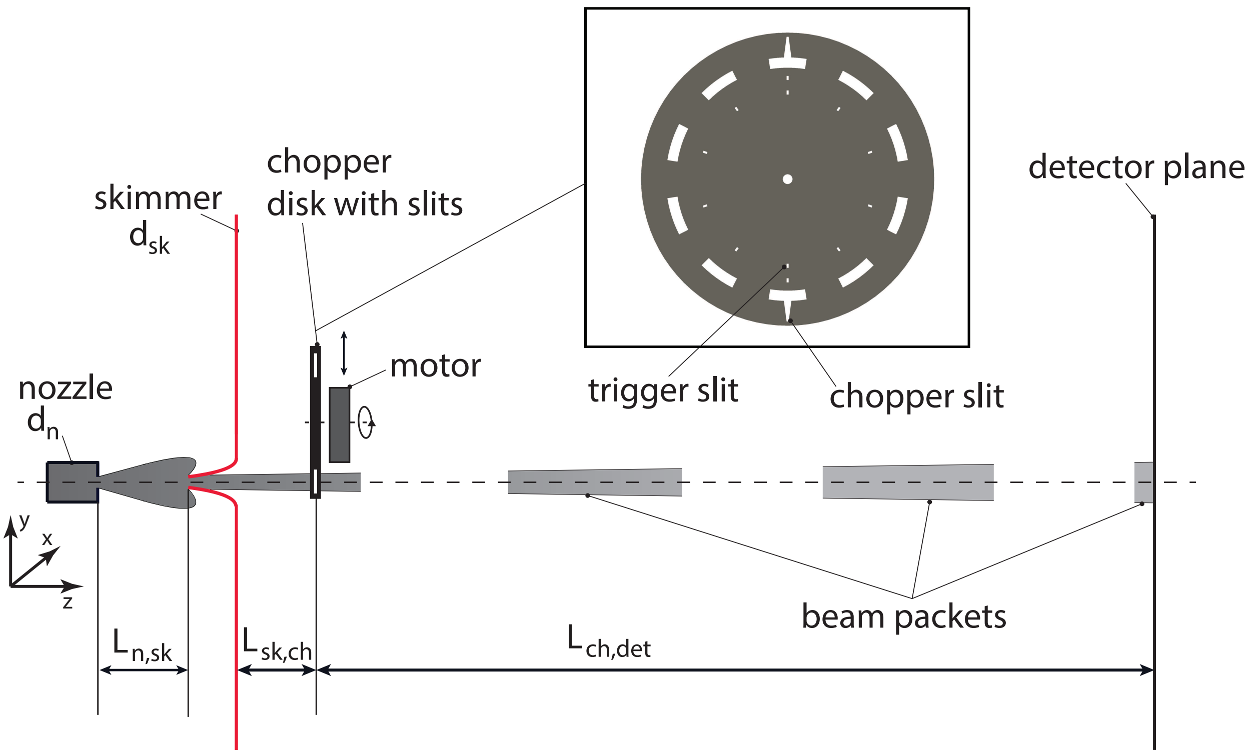

FIG. 1. Schematic representation of the TOF measurement set-up. Inset: Detailed drawing of the chopper disk with its two trapezoidal shaped slits placed 180° apart. The trigger slits are used to tag each beam packet. Further details are given in the text.

seen in Fig. 1.

The neutral helium beam was created by a free jet expansion from a source reservoir through a 10±1 µm diameter nozzle (Plano GmbH, A0300P). The central part of the beam was selected by either a standard skimmer (Beam Dynamics, inc.) with a diameter of 390 µm or with a self made glass microskimmer with a diameter of 4 µm. The microskimmer was made using a commercial micropipette puller (Narishige, PP-830) and led glass tubes (Corning 8161) with an outer diameter of 1.5 mm and an inner diameter of 1.1 mm. The key challenge when pulling microskimmers is to keep a relatively large opening angle even for the small apertures. Due to the somewhat manual nature of the skimmer pulling procedure, it is difficult to reproduce exactly the same openings and angles. Generally the best skimmers were obtained by using a relatively high heating settting (70), high pulling force (about 100 gr) and several heating steps (6). Decreasing the temperature or decreasing the number of heating steps made the taper of the skimmer longer and hence the opening angle smaller. Figure 3 shows a stereo microscope image (a) and a scanning electron microscope (SEM) image (b) of our self made skimmer. After pulling, the glass tube was glued onto a copper holder using two component glue (UHU PLUS ENDFEST 300). After the glue had hardened the glass tube was cut as short as possible to the inner rim of the copper holder using a diamond knife to just leave the top part. The mounting was done using a stereo microscope. Care was taken to ensure that the skimmer opening was parallel to the mounting base so that the beam and skimmer opening were perpendicular.

For all experiments the skimmer was placed 11.5±0.5 mm in front of the nozzle ($L_{n,sk}$). The distance from skimmer to choppper was 525±1 mm ($L_{sk,ch}$) and the distance from chopper to detector was 1905±5 mm ($L_{ch,det}$). The beam source in MAGIE has been specifically designed for microskimmer experiments, and is to our knowledge the only molecular beam source which allows positioning of the skimmer relative to the nozzle with sub-micrometer precision[23]. The source was operated at pressures in the range 11-181 bar at two different source temperatures, nominally 300 K and 125 K, obtained by cooling the nozzle with liquid nitrogen. For the alignment of the nozzle relative to the microskimmer the nozzle is moved in x and y direction across the skimmer opening (see Fig.1). The optimum nozzle to skimmer position is found when the detected beam signal reaches a maximum. The detailed alignment procedure can be found in Ref.[23]. Figure 2 shows a recorded 2D x/y scan intensity map for the alignment of the nozzle with the 4 μm diameter microskimmer for 4 different source pressure values of the 300K beam. As can be seen in Fig. 2 the spacial extension of the 2D source profile increases with increasing source pressure values. This corresponds well to the theoretical expected and experimentally verified behaviour of a spatial increase of the free jet expansion with pressure[25,26,28,29]. A higher source pressure leads to an increase in the detected source intensity likewise agreeing well with theoretical considerations. The most probable beam velocity and the beam velocity distribution were obtained by time of flight measurements (TOF). The beam was chopped by a mechanical chopper operated at frequencies of 310 Hz, 320 Hz and 230 Hz respectively. The chopper is linked to an optical diode which sends a trigger signal to the detector electronics so that the arrival time for the atoms in each beam pulse is recorded. The TOF signal is determined by the actual velocity distribution of the beam convoluted with the chopper slit and the detector function. When the velocity distribution is narrow (speed ratio high) it cannot be determined accurately using the standard deconvolution procedure described in[20]. We therefore used a new method recently developed in our group which allows the velocity distribution to be extracted with high accuracy[24].The modified method is based on a systematic variation of the chopper convolution parameters providing a set of independent

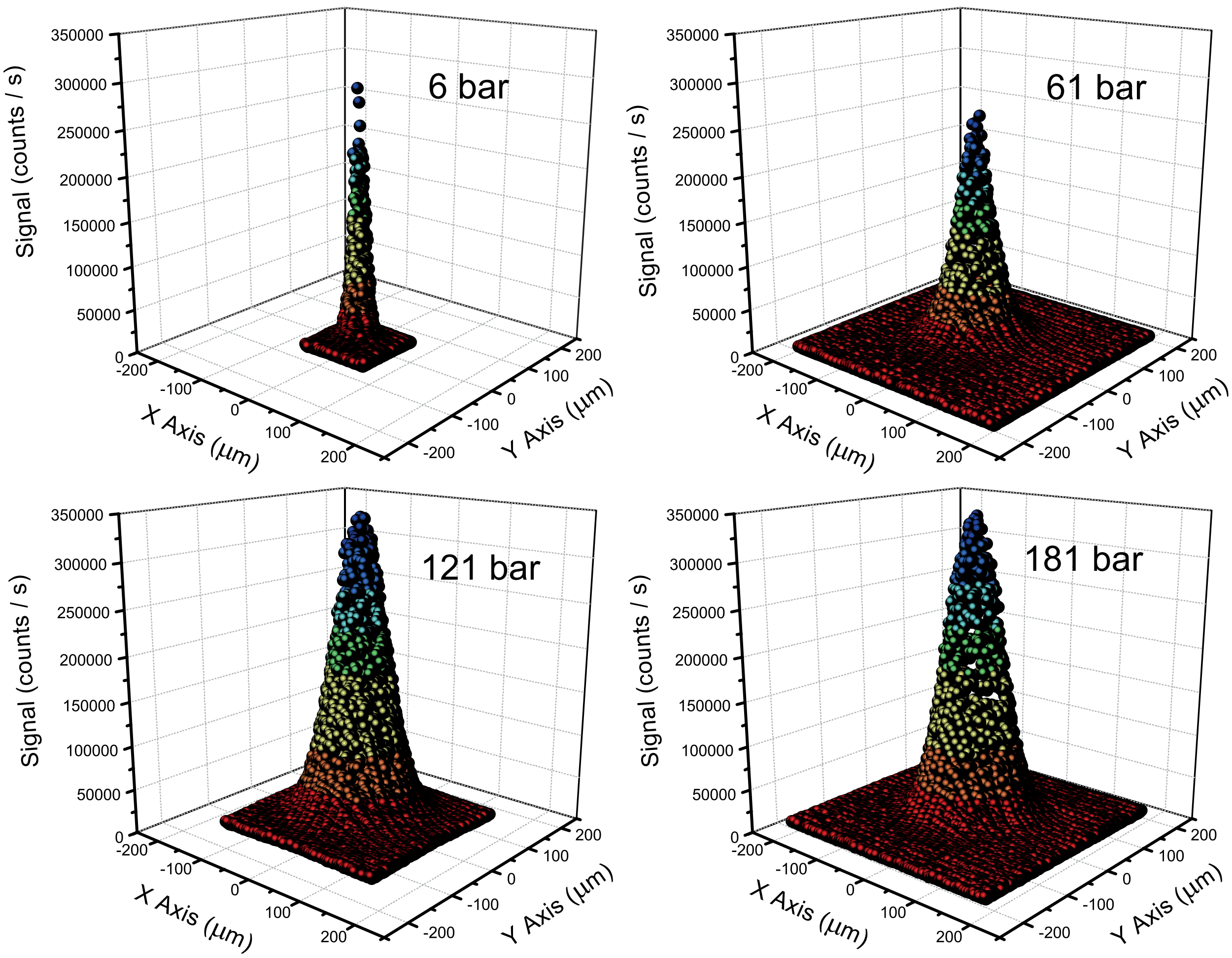

FIG. 2. 2D intensity maps recorded by scanning the 10 µm nozzle over the 4 µm glass skimmer. Since the glass skimmer diameter is small in relation to the spatial extension of the supersonic expansion this 2D intensity maps can be seen as an approximate image of the expansion itself.

measurements that can be fitted to obtain the helium beams speed ratio.

III. THEORETICAL MODEL

Our theoretical model for the supersonic helium expansion is based on a model proposed by Toennies and Winkelmann[30] in which the solution of the Boltzmann equation is obtained by means of the method of moments and assuming a Lennard-Jones (LJ) potential for the He-He interaction. The model was extended by Pedemonte et al.[31] to include other analytical He-He potentials, in particular the Hurly Moldover (HM) potential[32]. As in a previous work[26], the calculations presented were performed treating helium as a real gas and employing the equation of state obtained

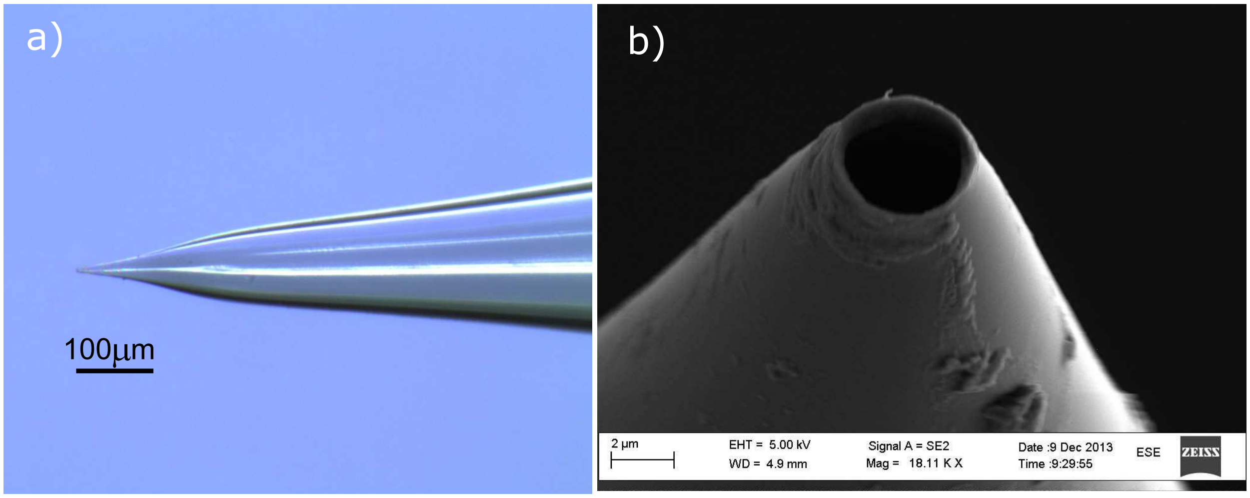

FIG. 3. (a) Stereo microscope image of the ∅ 4 µm microskimmer (glass). (b) SEM image of the ∅ 4 µm microskimmer (glass).

by McCarty[33].

The first assumption is to treat the expansion as spherically symmetric. Then an ellipsoidal velocity distribution, which consists of two Maxwell distributions parameterized by two different temperatures (denoted respectively $T_{||}$ and $T_{\perp}$ for the parallel and the perpendicular velocity components with respect to streamlines) is introduced:

$$f_{ell}(\vec{v}) = n\Big(\frac{m}{2\pi k_b T_{||}}\Big)^{\frac{1}{2}}\Big(\frac{m}{2\pi k_b T_{\perp}}\Big) \exp\Big(-\frac{m}{2k_b T_{||}}(v_{||}-u)^2 - \frac{m}{2k_b T_{\perp}}v_{\perp}^2\Big),$$

where m is the mass, n is the number density and u is the most probable velocity of the expanding gas. The evolution of the parameters $n, u, T_{||}$ and $T_{\perp}$ with the distance from the source z is obtained by solving numerically the equations which contain the collision integral $(2,1)$

$$\Omega^{(2,1)}(T_{eff}) = \Big(\frac{k_b T_{eff}}{\pi m}\Big)^{(1/2)} \int_0^{\infty} Q^{(2)}(E)\gamma^5 \exp(-\gamma^2) d\gamma,$$
$$\gamma = \sqrt{\frac{E}{k_b T_{eff}}} \quad , \tag{1}$$

where T_{eff} is an effective average temperature varying between $T_{\perp}$ and $T_{||}$, $Q^{(2)}$ is the viscosity cross section and E is the collision energy of two atoms in the center-of-mass system. For collisions between Bose-Einstein particles

$$Q^{(2)}(E) = \frac{8\pi\hbar^2}{mE} \sum_{l=0,2,4...} \frac{(l+1)(l+2)}{(2l+3)} \sin^2(\eta_{l+2} - \eta_l),$$

where η_l is the phase shift of the partial wave with orbital angular momentum l. For the present article, calculations were performed for LJ and HM potentials. Moreover we have also considered the Pirani et al. (PI) potential[34,35] which modifies and improves the LJ potential retaining a simple expression

$$V(r) = \varepsilon\Big(\frac{\mu}{n(r)-\mu}\big(\frac{r_m}{r}\big)^{n(r)} - \frac{n(r)}{n(r)-\mu}\big(\frac{r_m}{r}\big)^{\mu}\Big),$$

where for He, $\mu = 6$, r is the distance in the potential and $n(r)$ is given by

$$n(r) = \beta + 4\big(\frac{r}{r_m}\big)^2,$$

with parameters $r_m = 2.974$ Å, $\beta = 8$ and $\varepsilon = 2.974$ meV[36].

IV. RESULTS AND ANALYSIS

Fig. 4 and Fig. 5 show measurements of the most probable velocity (maximum velocity of the distribution) for different pressures for a cold and a room temperature beam. As can be seen there is good agreement between theory and experimental results, though we note that the velocities for the microskimmed beams are slightly lower (up to around 1%) for a given pressure for both temperatures. The reason for this is not quite clear. However, the smaller skimmer has a higher Knudsen number K_n, as can be found from Ref.[37] with $K_n = \lambda_0/d_{sk}$, where λ_0 is the mean free path of the helium atoms at the skimmer location and d_{sk} is the radius of the skimmer. A higher Knudsen number means a more fluid-like flow and it may be that this slows the beam down. It is strange though, that the effect does not increase with pressure.

Fig. 6 and Fig. 7 show the corresponding speed ratio plots for the two temperatures. The first thing to note is the near to perfect overlap between the microskimmer and standard skimmer measurements. Furthermore there is a reasonable agreement between theory and experiments, though it is interesting to see that for higher pressures the simulations seem to predict too high speed ratios for the cold beam and too low speed ratios for the warm beam. Comparing the three different potentials used for the simulation (LJ, HM and PI) the LJ potential gives the best agreement in the present experimental conditions (most prominent for the room temperature beam, see Fig. 7). This better agreement of LJ potential was also observed in[32] for temperatures above 50 K or in[25,26].

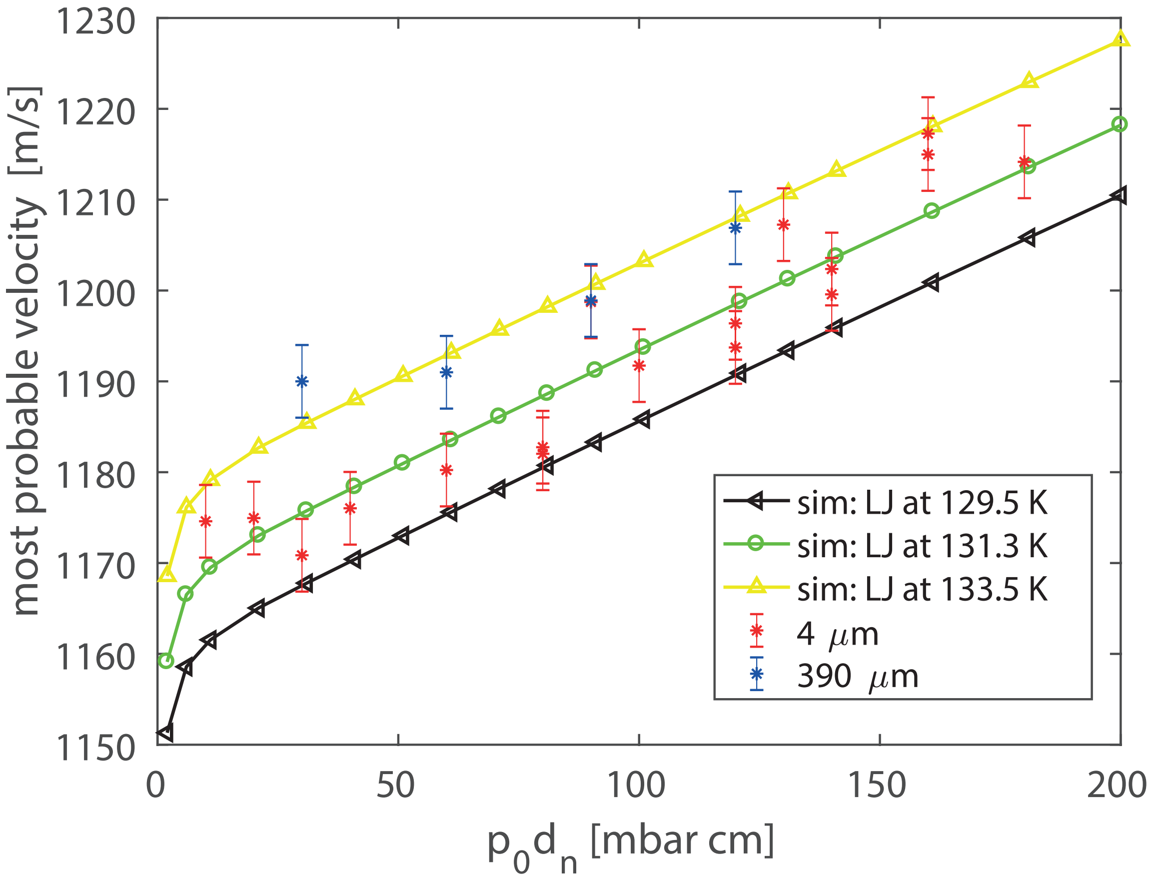

FIG. 4. Experimental results and simulations for the most probable velocity of cold beams as a function of p_0d, where p_0 is the source reservoir pressure and d_n the nozzle diameter. Note the slightly lower velocity for the microskimmer beam.

V. CONCLUSION

In this paper we have presented a systematic study of velocity distributions of helium beams collimated by a mikroskimmer for a room temperature beam and a cooled beam. The measurements were carried out in the pressure range 11 bar to 181 bar. Our results show that when the mikroskimmer is properly aligned with the nozzle, the speed ratio for the mikroskimmer does not differ from that of a standard skimmer. The most probable velocities for microskimmers appear to be slightly smaller than for standard skimmers. We measured a difference of up to around 1%. We contribute this to the higher Knudsen number for the microskimmer which leads to a more fluid like flow. Furthermore we show that the experimental data fit well to the theoretical model we have developed.

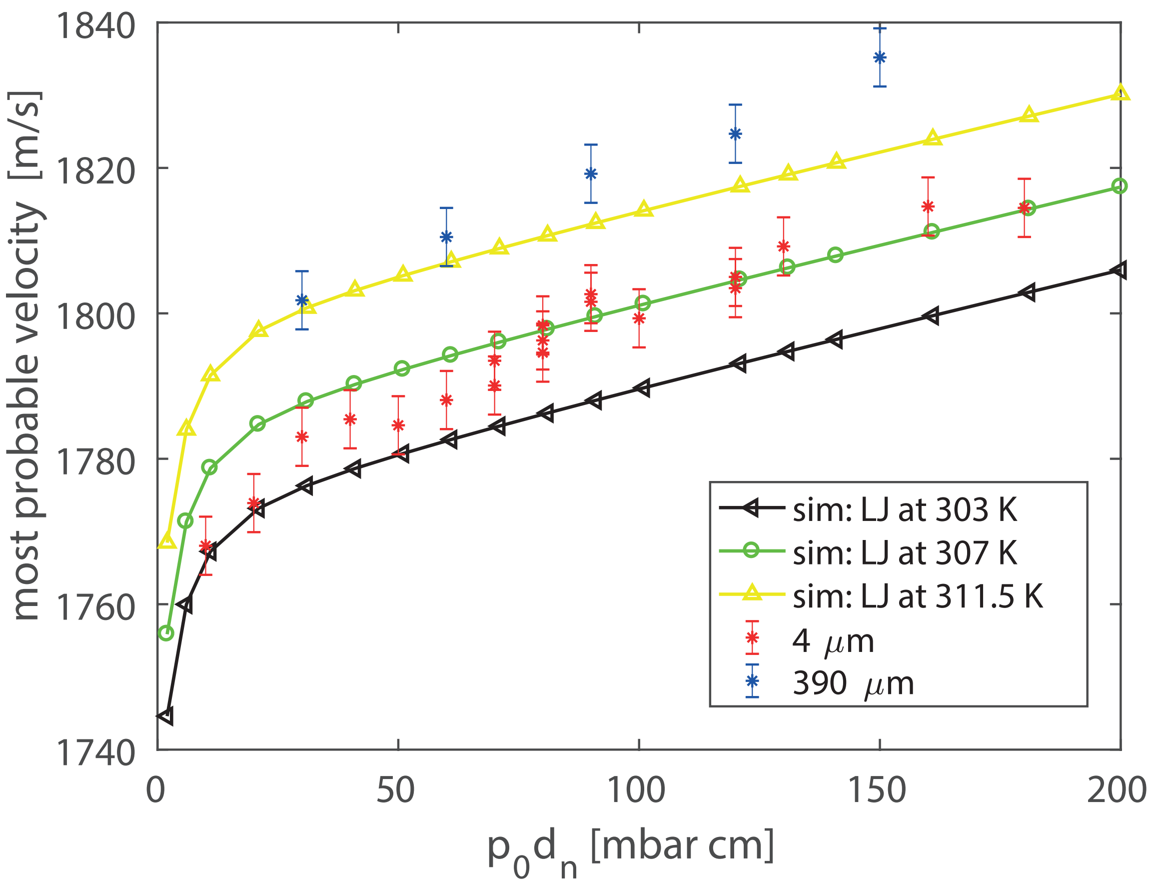

FIG. 5. Experimental results and simulations for the most probable velocity for room temperature beams as a function of p_0d, where p_0 is the source reservoir pressure and d_n the nozzle diameter. Note the slightly lower velocity for the microskimmer beam. This is discussed in the main text.

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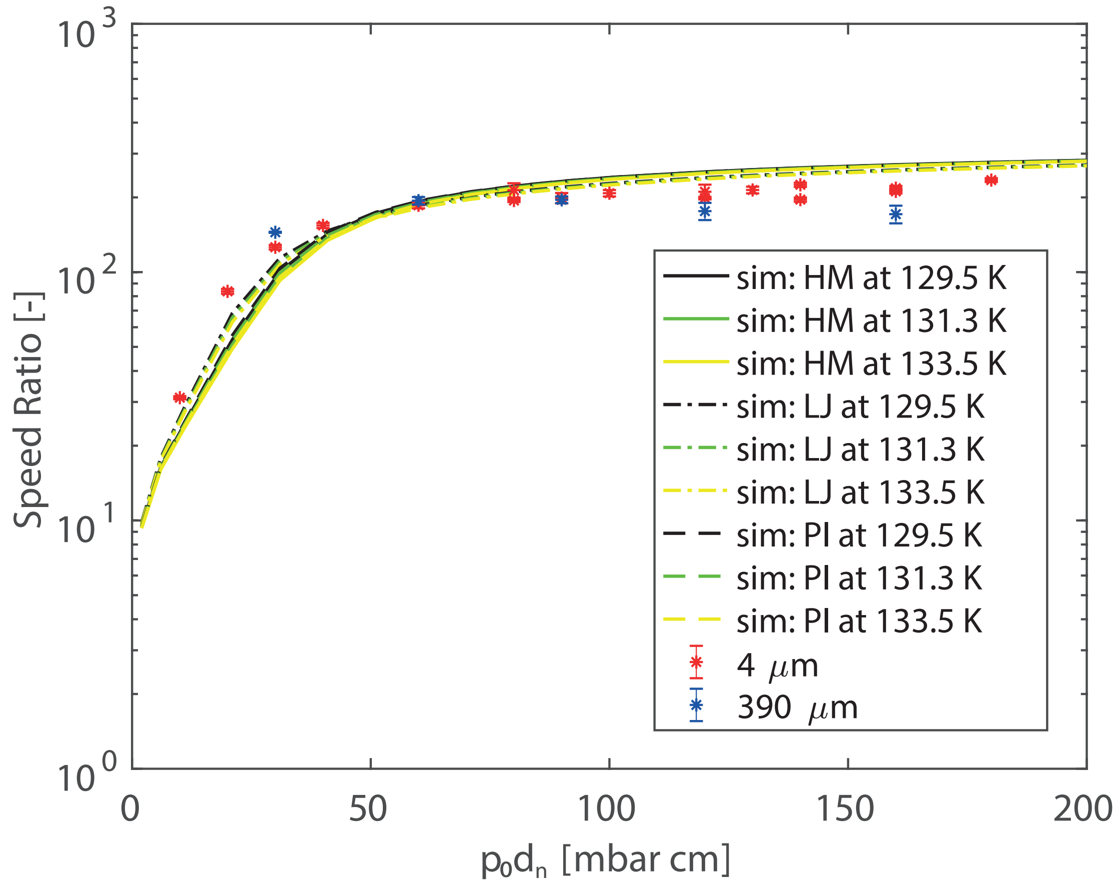

FIG. 6. Experimental results and simulations for the speed ratio of cold temperature beams plotted together with simulations. Note the very similar behaviour of microskimmer and standard skimmer. Note also the very little variations in the results for the different simulations at different temperatures.

REFERENCES

[1]D. Farías and K.-H. Rieder, Reports on Progress in Physics **61**, 1575 (1998).

[2]G. Bracco and B. Holst, "Surface science techniques," (Springer, 2013) Chap. 12, pp. 333–367, iSBN 978-3-642-34243-1.

[3]S. Eder, K. Fladischer, S. R. Yeandel, A. Lelarge, S. C. Parker, E. Søndergård, and B. Holst, Scientific Reports **5** (2015).

[4]B. Holst, M. Nohlen, K. Wandelt, and W. Allison, Phys. Rev. B **58**, R10195 (1998).

[5]D. Farías, H. Tröger, and K. Rieder, Surface Science **331-333, Part A**, 150 (1995).

[6]A. A. Taleb and D. Farías, Journal of Physics: Condensed Matter **28**, 103005 (2016).

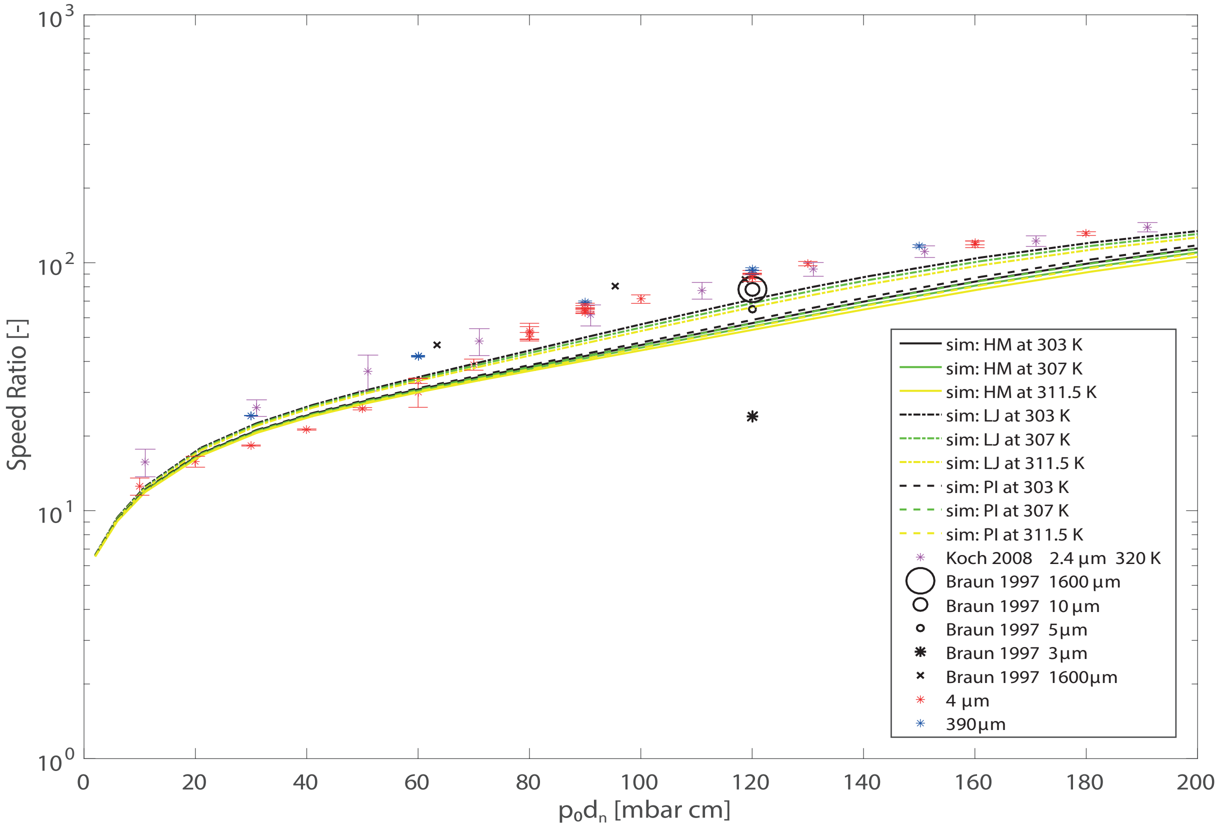

FIG. 7. Experimental results and simulations for the speed ratio of room temperature beams plotted together with simulations. For comparison, the speed ratio data from Braun et. al.[18] and Koch et. al.[11] are added to the plot. Note the very similar behaviour of microskimmer and standard skimmer.

[7] R. D. Brown, Z. M. Hund, D. Campi, L. E. OLeary, N. S. Lewis, M. Bernasconi, G. Benedek, and S. J. Sibener, The Journal of Chemical Physics **141**, 024702 (2014).

[8] J. Braun, P. Ruggerone, G. Zhang, J. P. Toennies, and G. Benedek, Phys. Rev. B **79**, 205423 (2009).

[9] A. A. Taleb, H. K. Yu, G. Anemone, D. Farías, and A. M. Wodtke, Carbon **95**, 731 (2015).

[10] C. Büchner, S. D. Eder, T. Nesse, D. Kuhness, P. Schlexer, G. Pacchioni, J. R. Manson, M. Heyde, B. Holst, and H.-J. Freund, accepted by Phys. Rev. Lett. (April 2018).

[11] M. Koch, S. Rehbein, G. Schmahl, T. Reisinger, G. Bracco, W. E. Ernst, and B. Holst, Journal of Microscopy **229**, 1 (2008).

[12] S. D. Eder, T. Reisinger, M. M. Greve, G. Bracco, and B. Holst, New Journal of Physics **14**, 073014 (2012).

[13]P. Witham and E. Sanchez, Review of Scientific Instruments **82**, 103705 (2011).

[14]A. Fahy, M. Barr, J. Martens, and P. C. Dastoor, Review of Scientific Instruments **86**, 023704 (2015).

[15]M. Barr, A. Fahy, J. Martens, A. P. Jardine, D. J. Ward, J. Ellis, W. Allison, and P. C. Dastoor, Nature Communications **7**, 10189 (2016).

[16]L. Vattuone, G. Bracco, M. Smerieri, L. Savio, and M. Rocca, "Dynamics of gas-surface interactions: Atomic-level understanding of scattering processes at surfaces," (Springer Heidelberg New York Dortrecht London, 2013) Chap. 1.

[17]Scoles, *Atomic and Molecular Beam Methodes*, edited by G. Scoles, Vol. 1 and 2 (Oxford University Press, New York Oxford, 1988).

[18]J. Braun, P. K. Day, J. P. Toennies, G. Witte, and E. Neher, Review of Scientific Instruments **68**, 3001 (1997).

[19]D. J. Auerbach, *Atomic and Molecular Beam Methodes*, edited by G. Scoles, Vol. 1 (Oxford University Press, 1988) pp. 362–379.

[20]H. Pauly, "Atom, molecule, and cluster beams 2," (Springer, 2000) Chap. 3.2, pp. 156–171.

[21]R. B. Doak, R. E. Grisenti, S. Rehbein, G. Schmahl, J. P. Toennies, and C. Wöll, Phys. Rev. Lett. **83**, 4229 (1999).

[22]T. Reisinger and B. Holst, J. Vac. Sci. Technol. B **26(6)**, 2374 (2008).

[23]S. D. Eder, B. Samelin, G. Bracco, K. Ansperger, and B. Holst, Review of Scientific Instruments **84**, 093303 (2013).

[24]A. S. Palau, S. D. Eder, T. Kaltenbacher, B. Samelin, G. Bracco, and B. Holst, Review of Scientific Instruments **87**, 023102 (2016).

[25]T. Reisinger, G. Bracco, S. Rehbein, G. Schmahl, W. E. Ernst, and B. Holst, The Journal of Physical Chemistry A **111**, 12620 (2007).

[26]S. D. Eder, G. Bracco, T. Kaltenbacher, and B. Holst, The Journal of Physical Chemistry A **118**, 4 (2014).

[27]A. Apfolter, *Wiederaufbau und Test einer He-Streuapparatur und erste Streuexperimente an amorpher sowie kristalliner SiO2-Oberflche*, Master's thesis, Graz University of Technology (2005).

[28]T. Reisinger, M. M. Greve, S. D. Eder, G. Bracco, and B. Holst, Phys. Rev. A **86**, 043804 (2012).

[29]T. Nesse, S. D. Eder, T. Kaltenbacher, J. O. Grepstad, I. Simonsen, and B. Holst, Phys. Rev. A **95**, 063618 (2017).

[30]J. Toennies and K. Winkelmann, The Journal of Chemical Physics **66**, 3965 (1977).

[31]L. Pedemonte, G. Bracco, and R. Tatarek, Phys. Rev. A **59**, 3084 (1999).

[32]L. Pedemonte and G. Bracco, The Journal of Chemical Physics **119**, 1433 (2003).

[33]R. D. McCarty and V. D. Arp, "A new wide range equation of state for helium. in: Advances in cryogenic engineering. advances in cryogenic engineering," (Springer, Boston, MA, 1990) pp. 1465–1475.

[34]F. Pirani, M. Albertí, A. Castro, M. M. Teixidor, and D. Cappelletti, Chemical Physics Letters **394**, 37 (2004).

[35]F. Pirani, S. Brizi, L. F. Roncaratti, P. Casavecchia, D. Cappelletti, and F. Vecchiocattivi, Phys. Chem. Chem. Phys. **10**, 5489 (2008).

[36]S. Longo, P. Diomede, A. Laricchiuta, G. Colonna, M. Capitelli, D. Ascenzi, M. Scotoni, P. Tosi, and F. Pirani, in *Computational Science and Its Applications – ICCSA 2008*, edited by O. Gervasi, B. Murgante, A. Laganà, D. Taniar, Y. Mun, and M. L. Gavrilova (Springer Berlin Heidelberg, Berlin, Heidelberg, 2008) pp. 1131–1140.

[37]G. A. Bird, Physics of Fluids **19**, 1486 (1976).